

# **Sulfuric Acid on Europa's Surface and the Radiolytic Sulfur Cycle**

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# Sulfuric Acid on Europa's Surface and the Radiolytic Sulfur Cycle

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Galileo infrared spectra of Europa's surface show distorted water bands that have been attributed to hydrated evaporite salts (McCord et al., J. Geophys. Res. 104, 11827, 1999) or to the scattering properties of ice (Dalton and Clark, Bull. Am. Astron. Soc. 30, 1081, 1998). Using new laboratory spectra, we show that hydrated sulfuric acid can explain Europa's spectra and further show that this species is product of radiolysis. Sulfuric acid on Europa occurs as the radiolytically stable octahydrate and hemihexahydrate and is a major surface component, along with water ice. The sulfuric acid concentration spatially correlates with Europa's visually dark material, which we identify as radiolytically altered sulfur polymers. Radiolysis by incident jovian plasma continuously cycles sulfur between three chemical reservoirs: sulfuric acid, polymerized sulfur, and sulfur dioxide, with the acid being about 50 times more abundant than the other forms because of the stability of the sulfate anion under irradiation. The original source of sulfur may be incident iogenic sulfur ions or endogenic sulfur compounds that are altered by radiolysis. Geological processes can bury and redistribute the sulfur compounds, producing a sulfur-rich crust and a non-uniform surface distribution. The low melting point of sulfuric acid and its ability to supercool may facilitate geological processes. Europa's magnetic response may be influenced by the electrical conductivity of sulfuric acid.

Europa is unique among Jupiter's moons. It is differentiated (1) with an icy crust that may melt from tidal heating to produce a subsurface ocean (2). The surface may be young and renewed by solid state convection (3) or extrusion of solid or liquid material from below (4). Europa's surface exhibits bright, icy plains and younger, darker mottled terrain (4). Linear features with different morphologies crisscross the surface. These long (>1000 km), narrow (10-20 km) features often show bright central bands flanked by bands of darker material ("triple bands"). Because Europa is within Jupiter's energetic magnetosphere, the surface is subject to intense bombardment by high-energy plasma ( $e^-$ ,  $H^+$ ,  $O^{n+}$ ,  $S^{n+}$ ) (5-7) that can alter the surface by radiolysis and ion implantation (8-10).

Europa's surface consists mainly of water ice and hydrated materials (11,12) with minor amounts of  $SO_2$ ,  $CO_2$ , and  $H_2O_2$  (9,10,13). Hydrated compounds on Europa were suggested by shifts and distortions of  $H_2O$  absorption bands found in spectra obtained by Galileo's near infrared mapping spectrometer (NIMS) (11,12). These spectra vary between two extremes (end-members), from ice-like, with a symmetric absorption band at 2- $\mu m$  in wavelength, to predominately hydrated with highly distorted  $H_2O$  bands (Fig. 1A). Most NIMS spectra of Europa's surface indicate mixtures of these two end-members. Although the hydrated material is ubiquitous on Europa, there is more of it within the linear features and the dark mottled terrain (12) of Europa's trailing (relative to orbital motion) hemisphere, which suffers the greatest plasma bombardment and implantation (5).

It was suggested (12) that Europa's spectra are similar to those of hydrated salt minerals (Fig. 1B) such as epsomite ( $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ ), magnesium hexahydrate ( $\text{MgSO}_4 \cdot 6\text{H}_2\text{O}$ ), and natron ( $\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$ ). Fits based on combining spectra of several salts were suggested to indicate a subsurface ocean (12), with salt minerals being formed on Europa's surface (14) by extrusion of subsurface brine that evaporates to form salt pans, similar to terrestrial deposits. However, identification of specific hydrates in the spectra is not unique (12).

The presence of  $\text{H}_2\text{O}_2$  and  $\text{SO}_2$  in the icy surface of Europa suggests that sulfuric acid ( $\text{H}_2\text{SO}_4$ ), a common photochemical product in the atmospheres of Earth and Venus, might be formed on Europa. Here we present laboratory spectra showing that NIMS observations of the European surface are consistent with the presence of hydrated  $\text{H}_2\text{SO}_4$ , and show that  $\text{H}_2\text{SO}_4$  is part of a radiolytic sulfur cycle.

Frozen  $\text{H}_2\text{SO}_4$  hydrates occur as  $\text{H}_2\text{SO}_4 \cdot n\text{H}_2\text{O}$ , where  $n = 1, 2, 3, 4, 6.5$ , and 8. (15-17). We investigated the  $n = 4, 6.5$ , and 8 hydrates by freezing stoichiometric solutions and obtaining their infrared (IR) reflectance spectra. The solutions contained spectrally neutral diamond powder to produce diffusely reflecting media. Samples with various grains sizes ( $\sim 5\text{-}\mu\text{m}$  to  $\sim 50\text{-}\mu\text{m}$  effective diameter) were prepared and spectra obtained for temperatures between  $\sim 80\text{ K}$  and their melting points ( $\sim 220\text{ K}$  to  $230\text{ K}$ ). The spectra (Fig. 1C) of  $\text{H}_2\text{SO}_4 \cdot 6.5\text{H}_2\text{O}$  and  $\text{H}_2\text{SO}_4 \cdot 8\text{H}_2\text{O}$  are similar, and show little variation with temperature. The hydration bands of  $\text{H}_2\text{SO}_4 \cdot 4\text{H}_2\text{O}$  were found to be less distinct than for the  $n = 6.5$  and 8 hydrates and unlike those

in Europa's spectra. Laboratory spectra of  $\text{H}_2\text{SO}_4 \cdot 8\text{H}_2\text{O}$  are similar to the spectrum of Europa's hydrated material (Fig. 1D) and provide the best match yet found for any single compound (18).

The small differences between the Europa and laboratory spectra (Fig. 1D) may be due to several effects. (i) The Europa spectrum also contains water ice features, inclusion of which may improve the comparison, particularly in the 1.2- $\mu\text{m}$  to 1.4- $\mu\text{m}$  region. (ii) The wavelengths of reflectance minima for hydration bands are reduced by ion irradiation and partial dehydration. Shifts of  $\sim 0.02 \mu\text{m}$  were produced by proton irradiation (19) and spectra of partially dehydrated sulfates exhibit shifts of  $0.04 \mu\text{m}$  (11). Such effects may be responsible for the small mismatch of band positions found in the Europa and laboratory spectra. (iii) The grain sizes of the laboratory samples may be different than those on Europa's surface. (iv) Other hydrates may also be present in Europa's spectrum.

Considering the above effects, the match to the NIMS spectra suggests that  $\text{H}_2\text{SO}_4 \cdot n\text{H}_2\text{O}$  ( $n = 6.5, 8$ ) is a major component of Europa's optically sensed surface. The optical sampling depth is a few grain diameters ( $\sim 60 \mu\text{m}$  on Europa, 9), so we adopt  $200 \mu\text{m}$  as the effective sampling depth. The number of  $\text{H}_2\text{SO}_4$  hydrated molecules for Europa's surface area whose spectrum is given in Fig. 1D is  $\sim 7 \times 10^{19} \text{ cm}^{-2} (200 \mu\text{m})^{-1}$ . The average number of  $\text{H}_2\text{SO}_4$  molecules for Europa's trailing hemisphere is about  $\sim 3 \times 10^{19} \text{ cm}^{-2} (200 \mu\text{m})^{-1}$ .

NIMS spectra of hydrated regions are featureless in the 3- $\mu\text{m}$  to 5- $\mu\text{m}$  region. We measured the mid-IR diffuse reflectance of a frozen  $\text{H}_2\text{SO}_4 \cdot 8\text{H}_2\text{O}$  sample and found no spectral features in this

range, consistent with NIMS spectra. We also measured the ultraviolet (UV) properties of  $\text{H}_2\text{SO}_4 \cdot 8\text{H}_2\text{O}$ . An absorption band was found at 190 nm with weaker absorption extending to 240 nm. A UV spectrum of Europa's trailing side (13) shows a possible absorption onset at 225 nm that could be due to  $\text{H}_2\text{SO}_4$  hydrate absorption. However,  $\text{SO}_2$  also has an absorption band beginning at these same wavelengths (20), and because a longer wavelength  $\text{SO}_2$  band appears in Europa's UV spectrum, the 225-nm onset could be due to  $\text{SO}_2$  alone. Therefore, Europa's UV spectrum is consistent with  $\text{H}_2\text{SO}_4$  hydrates existing on Europa.

The  $\text{H}_2\text{SO}_4$  concentration correlates with Europa's dark material.  $\text{H}_2\text{SO}_4$  hydrates are visually colorless, so the dark material is a related compound acting as a source, sink, or both. Sulfur has been suggested as a constituent for Europa's dark material (4, 21). We compare (Fig. 2) the visible reflectance of Europa's trailing side (22) to the reflectance of polymerized sulfur produced by radiolysis and photolysis (23-25). The shape of Europa's spectrum (approximately bilinear, with a breakpoint at 0.5  $\mu\text{m}$ ) is similar to the laboratory spectra and suggests that altered sulfur compounds are present on the surface. Because the visible reflectance of these materials and Europa are about the same, we estimate that the amount of polymerized S in Europa's optically sampled layer is  $\sim 4 \times 10^{17}$  S atoms  $\text{cm}^{-2}$  (200  $\mu\text{m}$ )<sup>-1</sup> (24,25). Therefore, we suggest three chemical reservoirs for Europa's sulfurous compounds: (i) polymerized sulfur, (ii)  $\text{SO}_2$ , with optically sensed surface densities of  $\sim 2 \times 10^{17}$   $\text{cm}^{-2}$  (200  $\mu\text{m}$ )<sup>-1</sup> (13), and (iii)  $\text{H}_2\text{SO}_4$  hydrate. Because extensive radiation dosage occurs to depths of 200  $\mu\text{m}$  in geologically short

times ( $\phi$ ), these reservoirs are in radiation-induced equilibrium, with sulfur being continuously cycled among these forms.

The production of  $\text{H}_2\text{SO}_4$  hydrate in this radiolytic cycle can be estimated using the  $G$ -value, the number of molecules produced per 100 eV of energy absorbed. Laboratory radiolysis of sulfur hydrosols produces sulfate (26) and irradiation of polymerized S in Europa's ice may be similar. For hydrosols,  $G(\text{SO}_4) \approx 0.6$  and applies to concentrations  $< 10^{-4}$  of the Europa's polymerized S concentration. However, these data apply to liquid suspensions and not frozen mixtures.

Irradiation of a 10:7 mixture of  $\text{H}_2\text{O}:\text{SO}_2$  ice produced compounds whose spectra are consistent with hydrated  $\text{H}_2\text{SO}_4$  (27). Radiolysis of  $\text{SO}_2$  ice produces sulfur, sulfate, and (predominately)  $\text{SO}_3$  (28). In the presence of water,  $\text{SO}_3$  rapidly forms  $\text{H}_2\text{SO}_4$  in an exothermic reaction (29). The  $G$ -value for  $\text{SO}_2$  in  $\text{H}_2\text{O}$  ice is not available so we use the value for  $\text{SO}_3$  production in pure  $\text{SO}_2$  ice ( $G \sim 5$ ) (28). With this  $G$  value, Europa's  $\text{SO}_2$  concentration, and using  $\sim 5 \times 10^{10} \text{ keV s}^{-1} \text{ cm}^{-2}$  for Europa's irradiation flux ( $\phi$ ), we obtain an  $\text{H}_2\text{SO}_4$  production of  $\sim 1 \times 10^9 \text{ s}^{-1} \text{ cm}^{-2}$ .

Irradiation of polymerized S in  $\text{H}_2\text{O}$  ice and associative reactions of  $\text{SO}_2$  with  $\text{H}_2\text{O}_2$  can also produce  $\text{H}_2\text{SO}_4$ , so the above rate is a lower limit. The optically observed  $\text{H}_2\text{SO}_4$  surface concentration [ $\sim 3 \times 10^{19} \text{ cm}^{-2} (200 \mu\text{m})^{-1}$ ] can be formed from  $\text{SO}_2$  and polymerized S in  $< 10^4$  years.

Sulfuric acid is quite stable under irradiation. When H is removed from  $\text{H}_2\text{SO}_4$  it recombines quickly (30). Destruction of  $\text{SO}_4$  is moderated by reactions between  $\text{SO}_2$  and  $\text{H}_2\text{O}_2$  molecules

produced during radiolysis (31). The net destruction rate of  $\text{H}_2\text{SO}_4$  can be estimated from rates for another acid sulfate,  $\text{Li}_2\text{SO}_4$ , which exhibits  $G \sim 10^{-3}$  to  $10^{-4}$  (32). These values are consistent with upper limits for  $\text{H}_2\text{SO}_4$  at Europa's concentration levels (31). The high radiation stability of  $\text{H}_2\text{SO}_4$ , particularly for the  $n = 6.5$  and 8 hydrates (31), implies large concentrations of  $\text{H}_2\text{SO}_4$  compared to  $\text{SO}_2$  and polymerized S. The observations show that Europa's  $\text{H}_2\text{SO}_4$  is more abundant than either  $\text{SO}_2$  or polymerized S by a factor of  $\sim 100$  (assuming that the photon sampling depths are about the same). The  $G$ -values, obtained for x-ray and  $\gamma$ -ray irradiation, give lower bounds for destruction by energetic particles of  $\sim 5 \times 10^7$  to  $5 \times 10^8 \text{ s}^{-1} \text{ cm}^{-2}$ . Heavy ions and UV radiation can also decompose  $\text{H}_2\text{SO}_4$  in the upper few  $\mu\text{m}$  of the surface (6) but  $G$ -values and cross-sections are not available.

The original source of sulfur may be  $\text{S}^{n+}$  ions implantated from the jovian plasma. The plasma influx,  $\sim 6 \times 10^7 \text{ S ions cm}^{-2} \text{ s}^{-1}$  (7), provides sufficient sulfur to account for the three reservoirs in  $10^4$  years. However, implantation would be expected to produce a more uniform surface distribution than is observed, so if the ultimate source of sulfur is plasma implantation, then some geological process has acted to produce the observed non-uniform distribution. One possibility is burial of implanted sulfur and its radiolytic products by solid state convection, forming a crust enriched with  $\text{H}_2\text{SO}_4$ ,  $\text{SO}_2$ , and S polymers. Subsequent convection of matter back to the surface may produce a non-uniform distribution of sulfurous material. Cracks in this crust can be filled by extrusion of ice or liquid water from below. This upwelling material can scour the crack walls, forming two layers of upward moving sulfurous till. The surface



expression would be two dark bands and possibly a bright median of pure ice, accounting for the appearance of Europa's triple bands (4).

Alternatively, much of the sulfurous material on Europa's surface may have been endogenic. Sulfuric acid formed internally could be geologically emplaced onto the surface. Extrusion of salt rich brine (12) may have provided sulfate salts to the surface, where proton implantation can replace salt mineral cations to form  $\text{H}_2\text{SO}_4$ . Radiolysis of such evaporite salt minerals should also produce  $\text{MgO}$ ,  $\text{MgOH}$ ,  $\text{Mg}(\text{OH})_2$ ,  $\text{Na}_2\text{O}$ , and  $\text{NaOH}$  (14, 33). However, hydroxides exhibit 1.4  $\mu\text{m}$ -bands (34) that are not evident in NIMS spectra (Fig. 1A) and imply hydroxide concentration limits of  $< 10\%$ .

The melting point of  $\text{H}_2\text{SO}_4$  solutions can be as much as 55 K lower than that of pure water ice (15), so liquid  $\text{H}_2\text{SO}_4$  may be formed more readily by tidal heating than melted brine. Enhanced  $\text{H}_2\text{SO}_4$  concentrations are associated with geological structures whose formation may have been affected by the presence of liquid  $\text{H}_2\text{SO}_4$ . Sulfuric acid can supercool to 150 K (16), possibly facilitating cryovolcanic activity. The acid is also an electrical conductor and may contribute to Europa's magnetic signature (35).

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## FIGURE CAPTIONS

Fig. 1. IR Reflectance spectra of Europa and candidate surface materials. (A) NIMS Europa end-member spectra, ice  $\gg$  hydrate (—) and hydrate  $\gg$  ice (—○—○—). (B) Evaporite salt minerals (12). (C) Spectra of  $\text{H}_2\text{SO}_4 \cdot n\text{H}_2\text{O}$  for various  $n$ , grain size  $d$ , and temperature  $T$ :  $n = 8$ ,  $d = 5 \mu\text{m}$ ,  $T = 80 \text{ K}$ , offset by 0.10 (— — —);  $n = 8$ ,  $d = 50 \mu\text{m}$ ,  $80 \text{ K}$ , offset by 0.05 (— — —);  $n = 8$ ,  $d = 50$ ,  $T = 140 \text{ K}$  (—);  $n = 6.5$ ,  $d = 50 \mu\text{m}$ ,  $T = 80 \text{ K}$  (.....). (D) Comparison of Europa hydrate spectrum (○○○, from A) and  $\text{H}_2\text{SO}_4 \cdot 8\text{H}_2\text{O}$  at  $T = 140 \text{ K}$ ,  $d = 50 \mu\text{m}$ , normalized at  $1.1 \mu\text{m}$  (—, from C).

Fig. 2. Reflectance spectra of Europa's trailing side compared to photolyzed and radiolyzed sulfur compounds. Europa spectrum (22) (● ● ● ●); Photolyzed  $\text{H}_2\text{S} + 10 \text{ H}_2\text{O}$  (23) (—); Transmission of ion irradiated  $\text{SO}_2$  (24), assumed here to approximate the reflectance when the same number of molecules are in the optical path (— —);  $\text{SO}_2^+$  irradiated  $\text{H}_2\text{O}$  ice (25), assumed to represent implanted and irradiated S in  $\text{H}_2\text{O}$  ice (—○—○—).



